

THREE DIMENSIONAL SIMULATION OF SPATIAL AND TEMPORAL VARIABILITY OF STRATOSPHERIC HYDROGEN CHLORIDE

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Abstract. Spatial and temporal variability of atmospheric HCl columns are calculated for January 1979 using a three-dimensional chemistry-transport model designed to provide the best possible representation of stratospheric transport. Large spatial and temporal variability of the HCl columns is shown to be correlated with lower stratospheric potential vorticity and thus to be of dynamical origin. Systematic longitudinal structure is correlated with planetary wave structure. These results can help place spatially and temporally isolated column and profile measurements in a regional and/or global perspective.

Introduction

Hydrogen chloride (HCl) is the most abundant chlorine-containing molecule formed in the stratosphere. It is a reservoir species for chlorine atoms formed from the breakdown of naturally occurring and anthropogenic Cl-containing source gases. The chlorine in HCl is not active in ozone destruction but HCl is converted by reaction with OH to catalytically active forms. Since amounts of chlorine-containing source gases such as the chlorofluorocarbons CFCl_3 and CF_2Cl_2 are known to be increasing [WMO, 1986], it is expected that amounts of stratospheric HCl should also be increasing.

Mankin and Coffey [1983] reported an increase in the HCl column using airborne measurements of infrared absorption. A much smaller trend was inferred on the basis of ground-based measurements from Jungfraujoch, Switzerland [Zander et al., 1987], but reanalysis of the Jungfraujoch data taking into account the seasonal variation of HCl suggested a larger increase [Ozone Trends Panel, 1988].

Determination of the trend in HCl from column data is complicated by its natural variability. For example, Marche and Meunier [1983] reported a 25% increase in the HCl column over the Observatoire de Haute Provence over a one-week period. Ground-based columns are affected by variability in tropospheric HCl, which makes up a non-negligible fraction of the total column and which is sensitive to the precipitation history of the air mass being observed [Vierkorn-Rudolph,

1984]. There is also seasonal variability and the suggestion of important longitudinal variability during periods of strong dynamical activity based on calculations performed with a three-dimensional stratospheric coupled chemistry-transport model [Kaye and Rood, 1989]. No global satellite data, which might validate these calculations, currently exist. Very limited data on the variability of HCl have been obtained with the shuttle-based Atmospheric Trace Molecule Spectroscopy (ATMOS) instrument [Raper et al., 1987].

In this paper we report a detailed simulation of the spatial and temporal variability of stratospheric HCl during January 1979 using a three-dimensional chemistry transport model [Rood et al., 1989]. The dynamics of the stratosphere in January 1979 were characterized by a planetary wave number one pulse on about 10 January and a minor warming late in the month. The wave one activity of 1979 is not atypical of wintertime conditions as is confirmed by the pronounced wave one signature in total ozone climatology [London, 1980].

The transport characteristics of the model in the lower stratosphere have been verified by comparison to Limb Infrared Monitor of the Stratosphere (LIMS) data from the Nimbus 7 satellite. Rood et al. [1989] have shown that HNO_3 transport is accurately represented at high latitudes. Recent calculations on O_3 show by comparison with data from the Total Ozone Mapping Spectrometer (TOMS) instrument on Nimbus-7 that its behavior below 10 mbar is properly modeled throughout January and February of 1979. Applications of the modeling technique for the period of the Airborne Arctic Stratosphere Expedition (January to mid-February 1989) suggest the model can represent local behavior of stratospheric constituents for periods of six weeks or longer. A paper detailing the HNO_3 and O_3 simulations for the winter of 1979 has been submitted for publication.

Attention is focused on the total atmospheric column of HCl above the ground and its dependence on longitude, latitude, and time. These are the first calculations of expected variabilities in HCl distribution using a ground-to-stratopause three-dimensional model and show that the variabilities are indeed important and can be related to dynamical activity in the lower stratosphere. To our knowledge this is the first exposition of the capabilities of a 3-D model with chemistry and realistic winds as an aid in the interpretation of isolated minor constituent measurements.

Model Calculations

The basic chemistry/transport model used is described in Rood et al. [1989] and only its crucial features are outlined here. The model solves the continuity equation for HCl including both chemical and advective processes using winds from a data assimilation procedure [see Rood et

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al., 1989 and references therein] and zonally averaged and time-independent chemistry parameterizations. Advection is calculated with the spectral method using rhomboidal 30 truncation. This corresponds to approximately 5° longitude by 4° latitude resolution. The model has 19 vertical levels in the altitude range from 0 - 55 km.

The initial HCl concentration (Figure 1) and the chemical production rate and loss coefficients are calculated using results from the two-dimensional model described by Douglass et al. [1989]. This model has been validated by extensive comparison with constituent data as well as with other two dimensional models [Jackman et al., 1989]. The total tropospheric mixing ratio of chlorine is 2.48 parts per billion by volume (ppbv), while the maximum stratospheric mixing ratio of free chlorine ($\text{Cl} + \text{ClO} + \text{HCl} + \text{ClONO}_2 + \text{HOCl}$) is 2.05 ppbv. The lower tropospheric mixing ratios of 0.05 ppbv are consistent with those measured by Vierkorn-Rudolph et al. [1984]. HCl production is assumed to occur from the reactions of Cl with CH_4 , HO_2 , and CH_3O , while loss of HCl is due to reaction with OH and rainout in the troposphere. Reaction rates are those of DeMore et al. [1987]. Rainout rates are those in the Goddard two-dimensional models [Douglass et al., 1989 and references therein] and are assumed to be zonally symmetric and constant in time. These assumptions may lead to decreased temporal variability in the tropospheric contribution to the HCl column over that found in the atmosphere. Diurnal averaging of the reaction rates was carried out using the formalism of Turco and Whitten [1978]; the correction factors are only important for the reaction of Cl with HO_2 because both are diurnally varying.

Since there are no satellite data for HCl with which to initialize the model, a zonally symmetric field was assumed as in our previous work [Kaye and Rood, 1989]. Calculations carried out for N_2O with different starting dates assumed

showed that most of the sensitivity to initial conditions is removed within about one week. We will thus focus our attention on the period of time after 8 days of integration. New initialization techniques based on potential temperature and potential vorticity mapping of 2-D model fields to the 3-D grid are being developed for our 1989 simulations.

Details of the numerical and computational aspects of the solution of the continuity equation have been described previously [Rood and Kaye, 1989].

Results and Discussion

The model results show spatial and temporal variability throughout the simulated period. Because the possibility of model drift is important, it is first necessary to assess the stability of the model, that is, whether the fields calculated differ in some monotonic way from the initial condition. In the region of interest at mid-northern latitudes, no such drift is obvious. For example, zonal mean profiles (not shown) have little change over a month at 58.5°N , with the possible exception of the lower troposphere where the chemistry is dominated by rainout. In particular, there is little change in the region of maximum number density between approximately 15 and 20 km. Thus, any changes at some particular longitude should reflect dynamical variability. A time-height series for HCl at 58.5°N and 7.5°E (near Stavanger, Norway) is shown in Figure 2 and indicates substantial variability in the 15-25 km region, especially a large pulse near day 10 and a more long-lived one near day 26. From day 8 to day 10 there is a nearly four-fold increase in the HCl mixing ratio at 91.5 mbar and this location, while from day 22 to 26 a greater-than two-fold increase is seen at 150, 91.5, and 53.1 mbar. Much smaller changes are seen in the troposphere and middle and upper stratosphere.

Spatial and temporal variability in the meridional dependence of the total HCl column is

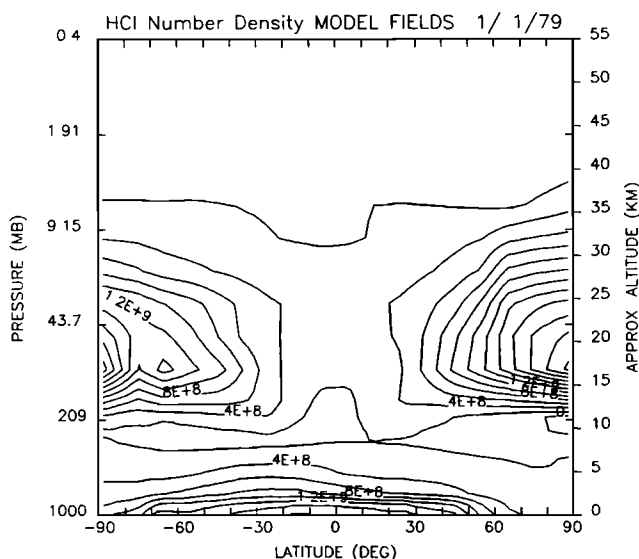


Fig. 1. Latitude-altitude plot of initial field of HCl used in chemistry-transport calculation obtained from "fast" two-dimensional model [Douglass et al., 1989] for 1979 chlorine levels.

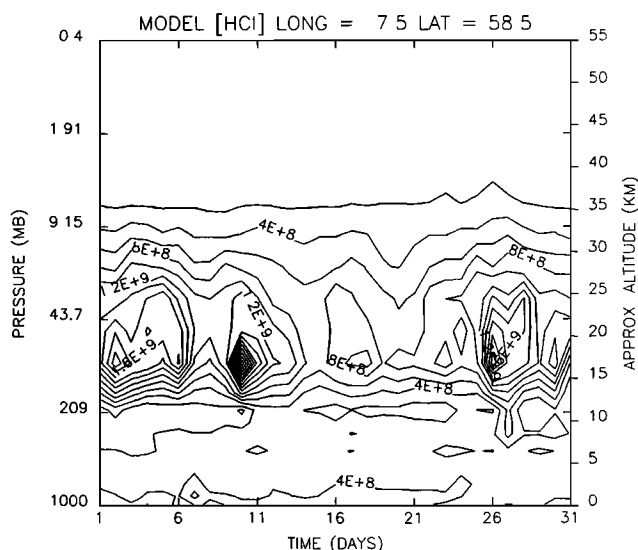


Fig. 2. Height-time series of HCl number densities at 58.5°N and 7.5°E .

presented in Figure 3, in which the columns are shown as a function of latitude for the initial field and at 10 day intervals through the calculation. Columns are shown both for at 0°E (bottom) and at 180°E (top). The zonal mean columns (not shown) show no substantial change with latitude over the course of the simulation other than a downward tendency at mid-latitudes in the northern and southern hemispheres, due mainly to loss in the troposphere. There is little variability in the tropics.

At 0°E there are some fundamental changes during the simulation. In particular, after 20

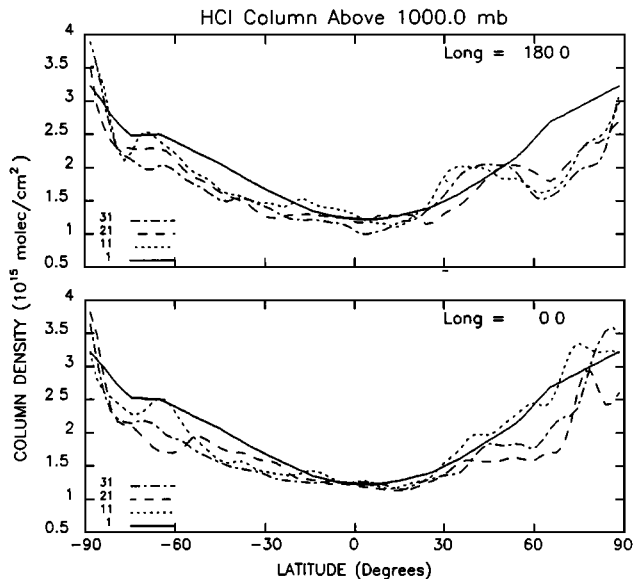


Fig. 3. Total HCl columns as a function of latitude for initial model day (solid line) and at ten day intervals throughout the calculation (line types indicated on figure). Bottom: 0°E; Top: 180°E.

and 30 days the smooth increase in column with latitude in the northern hemisphere seen in the initial field and after ten days has been replaced by a relatively flat region in mid-latitudes and a steep increase with latitude further north. This is indicative of the sharp edge of the polar vortex that develops during the warming. At 180°E, on the other hand, a minimum in the HCl column is seen near 60°N with sharply increasing columns as one approaches the pole. The minimum is due to isolation of the low HCl values in the Aleutian anti-cyclone. This pattern is established within ten days of the start of the integration and persists throughout the simulation. There is also much more variability in the tropics at this longitude than there is at 0°E.

Finally, the day-to-day variability of the HCl column at one longitude (7.5°E) for five latitudes (44.7°S, 1.1°N, 33.2°N, 58.5°N, and 79.1°N) is shown in Figure 4. It is seen that the smallest variability about the mean is in the tropics, being on the order of 10%, with day-to-day variability being substantially smaller. Larger variations are seen at 44.7°S, superimposed on a short-term downward drift during the first ten days or so of integration. Towards the end of the simulation period,

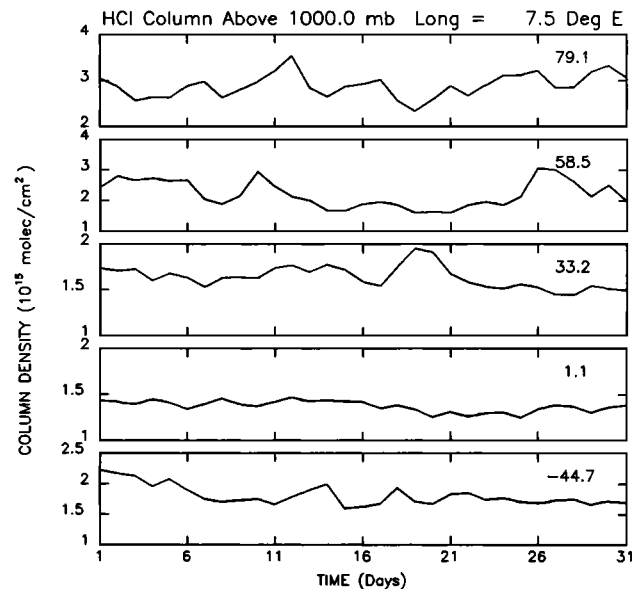


Fig. 4. Total HCl columns at 7.5°E vs. time at indicated latitudes: From top to bottom: 44.7°S, 1.1°N, 33.2°N, 58.5°N, 79.1°N.

however, the day-to-day variability at this point is quite small. At mid-northern latitudes, substantial day-to-day variability is seen. At 33.2°N there is a nearly 30% increase in HCl over a two day period (days 17-19) and a similar decrease over a 5 day period (days 19-24). Even larger percentage changes are observed at 58.5°N - a nearly 60% increase from day 8 to 10 and from day 24 - 26 (the period of the minor warming). The bulk of this variability is found to be in stratospheric HCl by considering only the HCl column above 200 mbar, which is close to the level from which the aircraft columns measurements are made.

The dynamical control of the HCl column is borne out by the high correlation of the HCl column with potential vorticity. This may be seen by comparing in Figure 5 the time series of

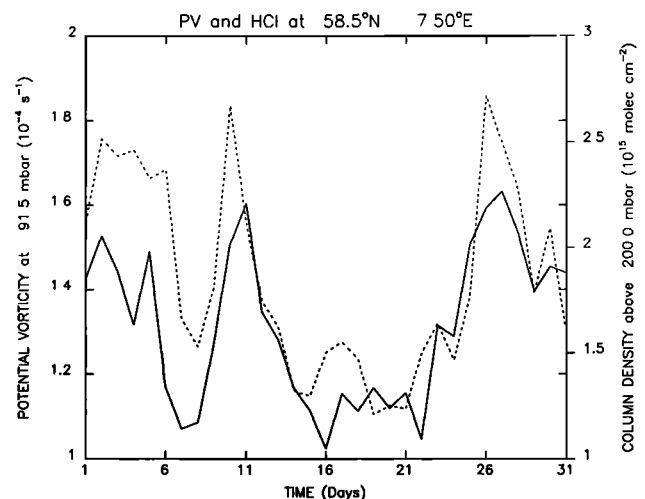


Fig. 5. Potential vorticity (solid line, left ordinate) vs. time at 58.5°N, 7.5°E, and 91.5 mbar and HCl column above 200 mb at same latitude and longitude (dotted line, right ordinate).

quasigeostrophic potential vorticity (solid line, left ordinate) at 7.5°E, 58.5°N, 91.5 mb and the corresponding HCl column above 200 mb (dotted line, right ordinate). The two peaks seen at days 10 and 26 in the HCl column are very clearly seen in the potential vorticity; the correlation coefficient between the total HCl column and potential vorticity at 91.5 mb is approximately 0.8. Similar correlation is also seen with the potential vorticity at 53 mb. Further evidence of dynamical origins comes from observation that the day 9-11 and 26-30 periods have the largest meridional flux at this location over the month of simulation.

Summary and Conclusions

The spatial and temporal variability of the atmospheric HCl column during winter has been characterized by use of a 3-D chemistry/transport model. Large (some 30-50% over several days) short-term variability is calculated and is shown to be related to HCl transport in the lower stratosphere. The variability is similar in magnitude to that which has been observed in ground-based measurements. Longitudinal asymmetry in the HCl columns associated with that in lower stratospheric planetary wave activity is also seen. These results suggest that extreme caution must be used in inferring monthly zonal average columns from limited numbers of measurements at times and locations of strong atmospheric dynamical forcing. This is especially true for long-term trend analysis, where the year-to-year increase in HCl (on the order of several percent per year) is much smaller than the short-term temporal variability (up to several tens of percent over a few days) presented here.

This paper reports on the first application of a realistic 3-D model to place isolated data in a global and regional context. Our success in simulating LIMS-observed HNO₃ and TOMS-observed O₃ give us confidence that the 3-D model is a useful tool for the applications considered here. We are currently extending the model to allow for parameterized heterogeneous loss of HCl and hope in the future to add an improved parameterization for tropospheric rainout.

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